THE EFFECT OF INSTRUMENTAL UNCERTAINTIES ON AFM INDENTATION MEASUREMENTS

Mark VanLandingham, Ph.D.
Materials Science Program and Center for Composite Materials
University of Delaware

Introduction

The accuracy and precision of the quantitative measurements made with various types of scanning probe microscopes (SPMs) can be limited by instrumentation error. The piezoelectric scanners that are utilized in SPMs control either the motion of the cantilever probe with respect to a stationary sample or the motion of the sample with respect to a stationary probe. While these scanners offer many advantageous characteristics that are critical to the performance of SPMs, they also exhibit several behaviors, e.g. hysteresis and creep, that introduce uncertainties in measurements. Also, the nonlinearities associated with the photodiode used in the optical lever detection system can detract from the accuracy of measurements.

Each particular type of SPM method is delineated by the type of tip-sample interactions that are used to create the image or measurement. For example, in atomic force microscopy or AFM, the generated images and measurements are based on the interatomic forces between the probe tip and the sample surface. In fact, the tip-surface interaction forces can be monitored by using the AFM in force mode. During force mode, the probe tip is first lowered into contact with the sample, then is indented into the surface, and finally is lifted off of the sample surface. Concurrently, a detection system measures the probe tip deflection. Our system, which is a Digital Instruments D3000 SPM, is equipped with an optical lever detection system, in which a laser beam is reflected off the top of the probe and onto a segmented photodiode. Deflection of the probe tip thus produces a change in the photodiode voltage, ΔV_t, which can be monitored as a function of the vertical displacement of the piezo actuator, Δz_p. A plot of ΔV_t versus Δz_p is termed a force curve, an example of which is shown in Figure 1. The slope of the contact or repulsive portion of the force curve, sometimes referred to as the sensitivity, is denoted by Σ and has units of V / nm.

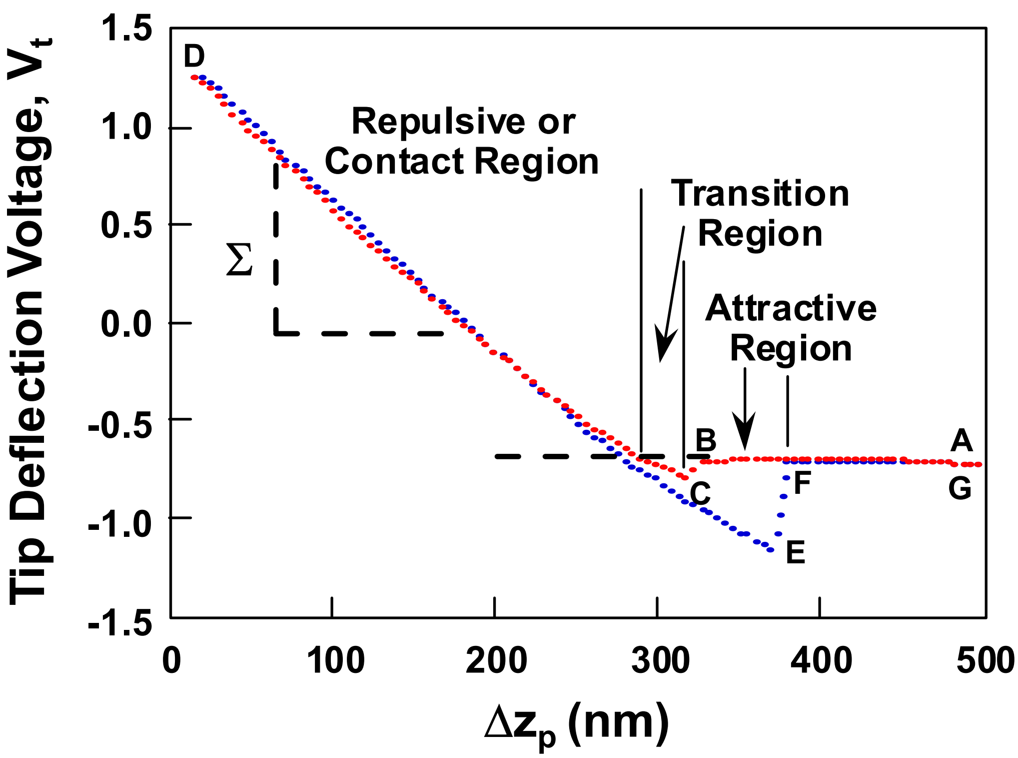


Figure 1 **AFM force curve of a polyurethane sample (E = 0.05 GPa) measured using a 3 N/m probe.**

Generally, force mode is used for minimizing tip-sample forces during imaging, but this mode can be used to configure the AFM to perform microscale and nanoscale indentation studies, as is discussed in more detail elsewhere [1-3]. The indentation displacement, Δz_i, is calculated by subtracting the displacement due to tip deflection, Δz_t, from the displacement of the piezoelectric actuator, Δz_p. The applied force can be calculated by multiplying Δz_t by the probe spring constant, k_c. For a sample which is infinitely stiff with respect to the probe, piezo motion will be translated directly into tip deflection, i.e., $\Delta z_p = \Delta z_t$. Thus, no indentation will occur and the slope of the contact portion of the force curve will reach a maximum value, Σ^*. The particular value of Σ^* is characteristic of a given probe and a given set of operating conditions and is extremely important in the calibration of the AFM indentation system and the subsequent determination of indentation response. In fact, for a sample that is deformed by the probe tip, Σ^* is used to calculate the relative amounts of indentation displacement and tip deflection. To ensure that accurate calibration is maintained throughout indentation testing, Σ^* values should be determined from force curve data for an "infinitely stiff" sample (e.g. sapphire) before and after indentation of the sample of interest.

In this article, the effects of instrumental uncertainties on AFM indentation measurements will be discussed. This discussion will include errors in indentation measurements due to (1) uncertainties in optical lever detection systems; (2) piezo nonlinearities; and (3) lateral motion of the probe tip that is due to bending of the cantilever.

Optical Lever Detection Systems

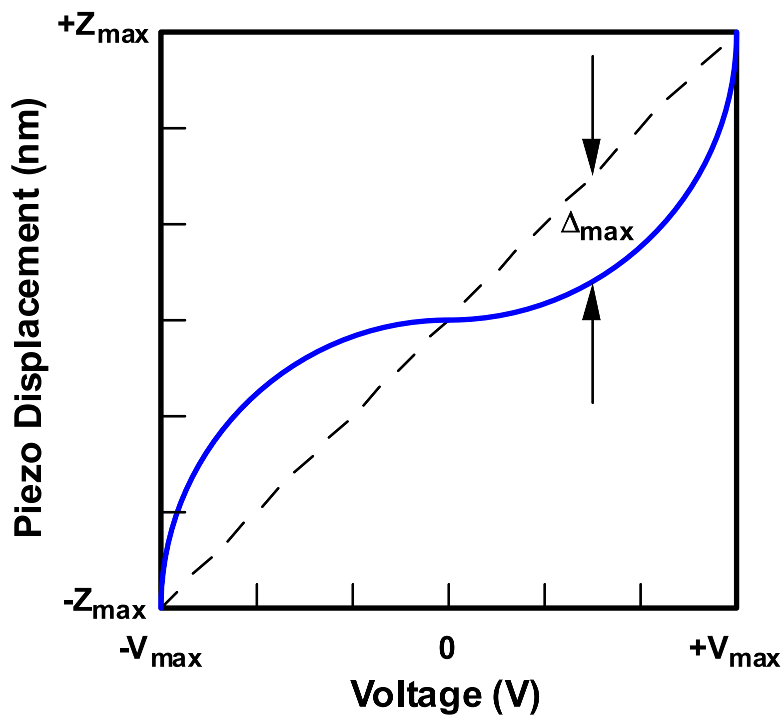
Reportedly, optical lever detection systems have noise levels on the subnanometer level [4], allowing for the excellent depth resolution of SPMs that employ such systems. While the precision of tip deflection measurements are on the order of 0.1 nm, the accuracy of such measurements could contain errors due to photodiode nonlinearities associated with the finite size of the laser spot. If the cantilever is deflected too far, the central portion of the spot no longer crosses the split in the photodiode. When the edge of the spot is crossing the split, a given spot movement produces less power, and therefore a lower sensitivity, than when the center of the spot crosses the split. For indentation measurements, significant nonlinearities in the tip deflection measurement will certainly lead to error and should be avoided. In order to produce small indents in polymer systems (E = 2-3 GPa) with stiff probes (k_c > 100 N/m), 50 nm or less of tip deflection has been sufficient. The corresponding voltage range of the photodiode used in our research is generally around ± 1 V, where ± 10 V represents the amplified range of the photodiode. In general, utilization of the center region of the photodiode will limit the errors due to photodiode nonlinearities to sufficiently small levels.

Piezo Nonlinearities

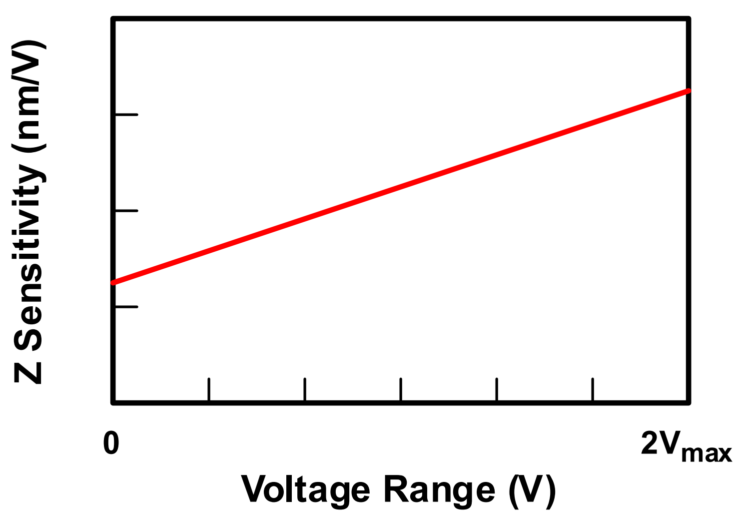
Several different types of nonlinearities exist in the operation of typical piezoelectric scanners utilized in SPMs. One type of nonlinear behavior is sometimes referred to as intrinsic nonlinearity. The relationship between applied voltage and piezo displacement is often approximated by a linear relationship over a certain range of voltage. The maximum deviation of the actual response from the linear curve fit, Δ_{max}, is often used to characterize this type of nonlinearity, as shown in Figure 2a. Also, for the z scanner, the coefficient relating the applied voltage range (e.g., from $-V_1$ to $+V_1$) and the resulting piezo displacement will be referred to as the z sensitivity, Z_s. Thus, $Z_s = \Delta z_p / 2V_1$. Because of the intrinsic nonlinearity, Z_s is a function of the applied voltage range, as shown in Figure 2b. Even for $\Delta_{max} / 2Z_{max} = 2\%$ (2-25% is the normal range for piezos used in AFM scanners [5]), a change in Z_s of more than 10% over the entire voltage range of the scanner can result [6]. This type of deviation is typical of so-called "hard" piezos, which is the type of piezoelectric material used in our z scanner. For indentation measurements, z motions in extension and retraction are often less than 300 nm, corresponding to voltage ranges of less than ± 10 V. Over a ± 10 V range, Z_s for hard piezo scanners typically varies by +/- 0.05 nm/V or less which translates directly into an error in z position of ± 1 nm.

Time-dependent piezo nonlinearities include aging and creep. Aging is a slow, gradual logarithmic decrease with time of the z sensitivity. In other words, later in its life, a scanner will not extend or contract to a given applied voltage as much as it did earlier in its life. Thus, the calibration of the scanner must be maintained, and a well-calibrated z scanner will help to minimize errors in indentation measurements. The calibration procedure for the z scanner that we follow on a regular basis involves a calibration sample with a known step height of 180 nm. Because the z motion during indentation measurements is of the same order, the errors caused by the slight aging between calibrations are estimated to be much less than 1 nm.

Creep, however, can be a much more problematic nonlinearity, at least with respect to making quantitative indentation measurements. Creep involves an initial displacement followed by a slow time-dependent displacement of the piezo caused by the application of an applied voltage. The time-dependent displacement typically occurs over a 10-100 second time interval [5]. By performing indentations using different force curve rates, the effects of creep can be observed.



(a)



(b)

Figure 2 **Illustration of intrinsic nonlinearity in piezoelectric materials: (a) maximum deviation of the actual response from the linear curve fit; and (b) z sensitivity as a function of the applied voltage range.**

To evaluate the effects of creep, force curves were acquired with different force curve rates. Σ^* values were measured for both extending and retracting (loading and unloading) piezo motions. Several different probes were used with a sapphire sample, and the force curve rate was varied from 8 Hz down to 0.01 Hz while all other indentation parameters were held constant. The extending and retracting Σ^* values remained approximately constant and equal down from 8 Hz to around 1 Hz, at which point they started to deviate, with extremely large differences at 0.01 Hz, as shown in Figure 3. This result was explained by the creep behavior of the piezo, which caused significant errors in Σ^* for rates of 1 Hz or less. Because of these results, all subsequent indentation measurements were made using force curve rates of 2 Hz or higher. This practice seems to limit errors caused by creep to acceptably small levels. As a result, Σ^* values and corresponding indentation data has been quite reproducible, whereas prior to the adoption of this practice, significant errors were prevalent.

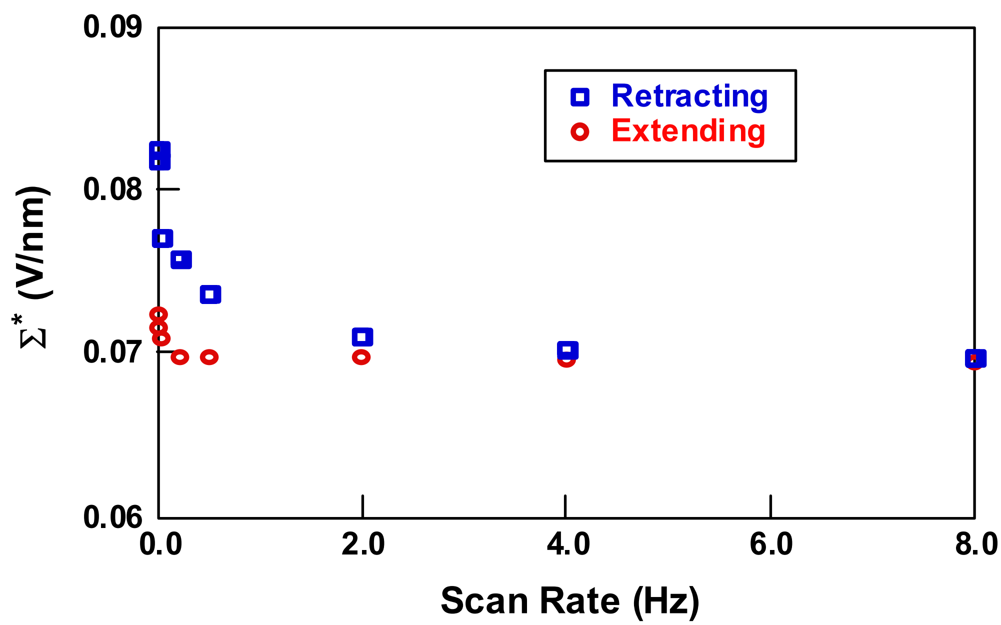


Figure 3 Σ^* **for extending and retracting curves as a function of the force curve scan rate for an S probe and a sapphire sample.**

While piezo nonlinearities are sometimes referred to as hysteresis, hysteresis is only one type of nonlinearity. If the voltage applied to a piezoelectric scanner is increased to some value and then decreased back to zero, the retraction of the piezo will not follow the same path as the extension of the piezo. This effect is called hysteresis and is illustrated in Figure 4. During scanning, hysteresis effects are often minimized by applying different voltage waveforms in the two directions. Currently, this type of correction is not available for the z scanner. However, hard piezo materials, such as that used for our z scanner, have been characterized by 4% hysteresis for ± 220 V, 2% for ± 110 V, and 1% for ± 50 V [6]. Thus for voltage ranges of ± 10 V or less used in indentation measurements, hysteresis will be on the order of 0.3%. Therefore, the errors incurred due to hysteresis will be very small, i.e., less than or on the order of ± 1 nm.

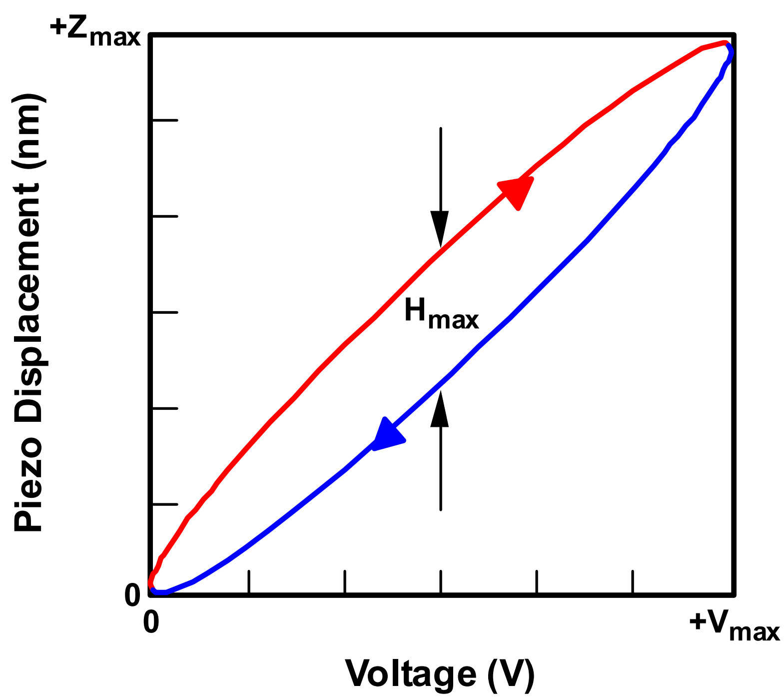


Figure 4 Illustration of hysteresis in piezoelectric materials. The upper curve represents piezo extension and the lower curve represents piezo retraction.

The final type of piezo nonlinearity that could effect indentation measurements is cross coupling. This term refers to movements in the x and y directions concurrent to the movement in the z direction during indentation. The x-z cross coupling is quite small compared to the lateral motion of the tip in the x direction that is due to the bending of the cantilever probe (discussing in the section to follow). Further, the compensation used to combat the lateral motion of the tip can also correct for the x-z cross coupling. The y-z cross coupling, however, cannot be corrected currently. Cross coupling effects would cause noticeable asymmetries in the plastic impressions created during indentation and nonlinearity in the force curves acquired using an infinitely stiff sample. For experiments in which all other sources of error have been limited, effects due to cross coupling have not been observed in our studies.

Lateral Motion of the Tip

As a cantilever beam bends, the angle of the beam at the free end will change. A probe tip attached to that free end will rotate and the end of the tip will translate laterally. The 10° angle of the probe tip to the x-y plane will add to this effect. For a tip in contact with a sample surface, friction, local deformation, and/or topography generally will restrict this lateral motion, and a lateral surface force will be generated that bends the cantilever in the direction opposing the bend due to the normal or indentation force. On a force curve plot, this effect will result in a decrease in the slope with increasing tip deflection. A lateral translation, Δx_p, of the probe that is proportional to the vertical translation, Δz_p, during indentation testing would counteract this effect, such that the measured deflection of the probe tip is due only to the surface normal force. Further, because the angle of beam deflection at the tip is on the order of 0.1°, a linear proportionality between the lateral compensating translation and the vertical displacement seems appropriate. To reduce the lateral motion effects inherent in our AFM system, new software was developed by Digital Instruments [7] to provide such a compensating lateral motion. This compensation attempts to counteract the moment acting on the cantilever due to the reaction forces at the surface.

For a particular probe-sample combination, increasing the amount of lateral compensation will decrease the size of the deformed region until an optimum range of compensation is found. This effect is shown in Figure 5 for an epoxy sample indented with a diamond-tipped stainless steel probe that is characterized by a spring constant of approximately 150 N/m. In this figure, the compensation angle, $\gamma = \tan^{-1} (\Delta x_p / \Delta z_p)$, for each indent is equal to 0°, 10°, 20°, 30°, 35°, 40°, 45°, and 50°, respectively, from left to right across the image. All other indentation parameters are constant. For $\gamma < 35°$, the indent area is larger than for $\gamma \geq 35°$ because the lateral translation of the tip causes it to push through the material to the left as Δz_p increases. For γ values of 45° and 50°, overcompensation has occurred, and a small amount of pile-up on the right side of the indent becomes apparent and is worse for $\gamma = 50°$. Thus, for γ between 35° and 40°, the motion of the tip is approximately vertical with respect to the local sample topography throughout the indentation process. This optimum range for γ correlates well with results of analytical and finite element models based on probe geometry. This correlation indicates that, for our system, x-z coupling is apparently a very small effect compared to the lateral motion that is related to the probe geometry during indentation.

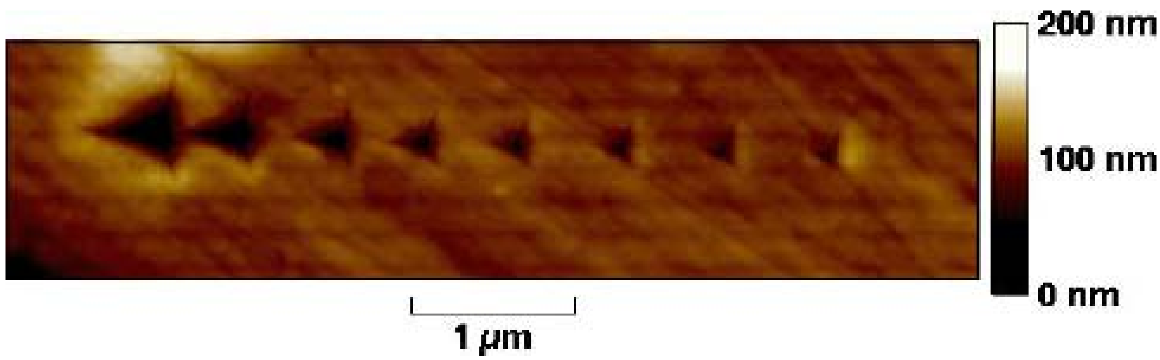


Figure 5 **AFM image of indents made with increasing lateral compensation angle, γ. From left to right, consecutive indents were made with γ = 0°, 10°, 20°, 30°, 35°, 40°, 45°, and 50°, respectively. Height scale from black to white is 0 to 200 nm.**

Evaluation of the indent areas produced on a relatively soft sample with different levels of compensation can yield an optimum compensation range for a particular probe. Narrowing that range to a precise optimum value requires indentation testing on a flat "infinitely stiff" surface (we have used a sapphire substrate for our work). As discussed previously, the slope of the force curve for such a sample will reach a maximum value, Σ^*, which can be calculated using a linear fit to the contact portion of the curve. Note that the relationship between ΔV_t and Δz_p is indeed linear for an infinitely stiff sample. Using simple mechanics expressions, this relationship is given by

$$\Delta V_t = \left(\frac{3C_\theta}{2L_c \cos(10^\circ)} \right) \Delta z_t = \Sigma * \Delta z_t$$

Thus, the linear fit is justified, and the scatter of the individual points about the fitted curve is assumed to be caused by the experimental uncertainties, such as those discussed in the previous sections.

In summary, for an optimum amount of compensation, the force curves on infinitely stiff samples will be linear, and the area of the plastic indents on relatively soft samples will be minimized with equivalent amounts of pileup on each side of the indent. We have found that the largest source of experimental uncertainty is caused by incorrect lateral compensation; too little compensation will result in the slope of the force curve decreasing with increasing tip deflection; too much compensation will result in the slope of the force curve increasing with increasing tip deflection. Therefore, to reduce the error in Σ^*, an optimum value of compensation must be used that results in only small deviations of data about the linear fit; e.g., deviations of less than ± 1 nm for 50 nm of tip deflection. Further, the values of Σ^* for a large number of consecutively measured force curves should be reproducible; e.g., a standard deviation of less than 5% of the average Σ^* value as calculated from 10-20 measurements of Σ^*. Even for optimum compensation, scatter of data points about the linear curve fits and scatter in Σ^* values about and average value will remain. These errors, possibly due to piezo or photodiode nonlinearities, will then propagate through calculations of Δz_t and Δz_i, which are used to characterize the indentation response. The significance of the errors with respect to the calculated indentation response, or to the differences in response of two different samples, can be estimated using standard error analysis techniques.

Summary and Final Comments

The AFM is certainly capable of performing microscale and nanoscale indentation measurements on a quantitative level. By quantitative, I mean that indentation response can be characterized by accurate and precise numerical values as long as steps, such as those discussed in this article, are taken to understand and limit experimental uncertainties. However, these numerical values cannot be used to calculate absolute properties of a sample. Only relative comparisons between sample responses can be made. This limitation is representative of indentation testing as a whole and is not specific to AFM indentation. It arises due to the complicated stress states and resulting deformation mechanisms associated with applying load to an extremely small area. For example, a hardness value is generally considered a quantitative measurement but can only be used for relative comparisons between materials. While hardness values can sometimes correspond to material properties, hardness itself is not a material property. Rather, hardness is a function of the loading geometry and calculations of hardness values do not consider the stress states or deformation mechanisms associated with indentation. Because of this relative nature of indentation results, the errors in the determination of the probe spring constant, k_c, are not important, because k_c is just a proportionality constant relating load to tip deflection and will cancel out in such relative comparisons.

Because the motion of the z piezo and the deflection of the probe tip during indentation tests is quite small compared to the total ranges of motion, errors due to photodiode and piezo nonlinearities that can be large over large ranges of motion are often small for the motions used during indentation tests. For creep, the rate of indentation testing is more important than the actual z motion, and thus force curve rates greater than 1 Hz are necessary to reduce errors due to creep. Also, use of the center region of the photodiode is recommended to limit photodiode nonlinearities. Even with these limitations on rates and motions, the AFM indentation technique cannot succeed without the use of lateral motion compensation. This compensation is critical to maintaining a force that is normal to the sample surface during an indentation event. Errors due to inappropriate compensation, photodiode nonlinearities, or piezo nonlinearities will lead to errors in the determination of Σ^*, which acts as a calibration parameter for the AFM indentation system. These errors will then propagate through the calculations of tip deflection and indentation displacement. As long as the errors are not significant with respect to the relative comparisons that are made between samples, this technique is a viable option for the characterization of microscale or even nanoscale indentation response of material systems.

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